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# EFFICIENCY OF SEPARATION OF LOW CONCENTRATION GASEOUS COMPONENTS BY COUNTER FLOW WASHING

Sándor SZENTGYÖRGYI, Károly MOLNÁR and Mária ÖRVÖS

Dept. of Chemical and Food Engineering
Technical University of Budapest
H-1521 Budapest, Hungary

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## Abstract

From the point of view of environmental protection, it is important to lower the sulphur dioxide emission of electric power plants and other industrial establishments. In our study, we have examined the factors affecting the efficiency of separating a given 'A' component from the gas phase, in the case of physical and chemical absorption techniques. We compared our conclusions with data obtained on a semi- industrial scale.

*Keywords:* gas-absorption, efficiency, wet washing.

## Introduction

When gases come in contact with liquids, the latter absorb some of the components of the gaseous system. During the contact of gases and liquids, two phenomena happen almost simultaneously: the diffusion of the gas into the mass of the liquid, which is accompanied by the vaporization of the liquid. The degree of diffusion is a characteristic of the gas–liquid system and the phase contact. The absorption of gases in liquids, when the gaseous component does not react with the liquid phase, is called physical absorption. If the absorption is accompanied by a chemical reaction, then the process is called chemosorption. In such cases the absorbed component reacts with the liquid phase, thus giving birth to a new component.

## Physical Absorption

Based on [1, 2] and *Figs. 1* and *2,* we may write that the diffusion velocity of component 'A' in the gas and liquid phases, respectively, is:

$$N_A = k_{AG}(y_{AG} - y_{Aj}) = k_{AL}(x_{Aj} - x_{AL})\,. \tag{1}$$

From *Fig. 3,* it can be seen that using the Henry constant we get

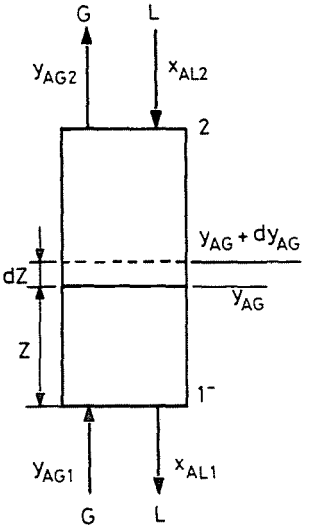


*Fig. 1.* Model of the absorption column

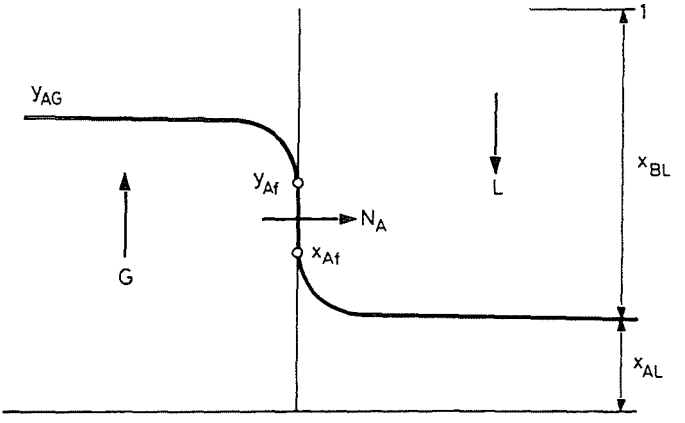


*Fig. 2.* The concentration distribution between the phases

$$y_{Af} = mx_{Af} \tag{2}$$

and from this follows that we can write:

$$N_A \left( \frac{1}{k_{AG}} + \frac{m}{k_{AL}} \right) = y_{AG} - mx_{AL} \,. \tag{3}$$

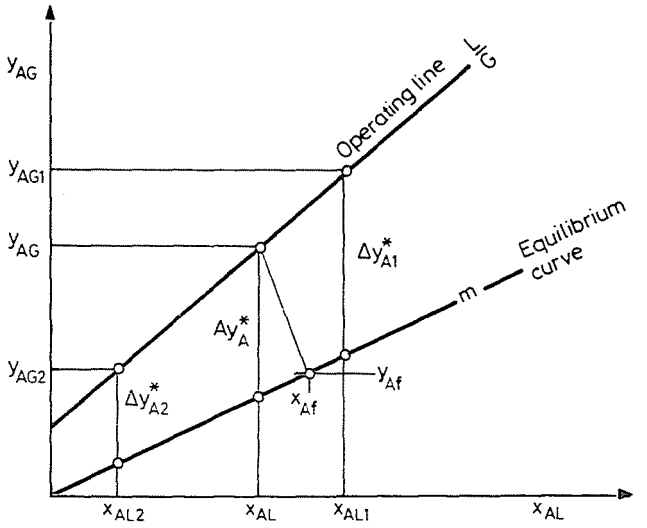


*Fig. 3.* Working line and equilibrium curve in the case of low concentration of the 'A' component

The overall mass transfer factor is:

$$K_y = \frac{1}{\frac{1}{k_{AG}} + \frac{m}{k_{AL}}}, \tag{4}$$

thus, the right side of *Eq.* (3) can be written:

$$\Delta y^*_A = y_{AG} - m x_{AL}. \tag{5}$$

From *Eqs.* (3), (4) and (5), the mass transfer velocity of component 'A' from the gas and into the liquid phase can be written:

$$N_A = K_y \Delta y^*_A. \tag{6}$$

From *Figs. 1* and *2* can be seen that for the elementary height of the column, the material balance for the gas side becomes:

$$N_A \mathrm{d}A_f = -G A_q \mathrm{d}y_{AG}, \tag{7}$$

where:

$$\mathrm{d}A_f = A_q a \mathrm{d}Z. \tag{8}$$

From here we can express the height $Z$ of the column:

$$Z = \int_0^Z \mathrm{d}Z = -\frac{G}{a} \int_{y_{AG1}}^{y_{AG2}} \frac{\mathrm{d}y_{AG}}{N_A}$$

and taking into account *Eq.* (6) we can write:

$$Z = -\frac{G}{K_y a} \int_{y_{AG1}}^{y_{AG2}} \frac{\mathrm{d}y_{AG}}{\Delta y_A^*} . \tag{9}$$

Expressed with gas phase driving forces, the fundamental equation of the washing tower can be written:

$$Z = H_{OG} \cdot N_{OG} \tag{10}$$

where

$$H_{OG} = \frac{G}{K_y a} \tag{11}$$

represents the height of transfer units, and

$$N_{OG} = \int_{y_{AG2}}^{y_{AG1}} \frac{\mathrm{d}y_{AG}}{\Delta y_A^*} \tag{12}$$

is the number of transfer units.

The efficiency has been defined in the following manner:

$$\eta = \frac{y_{AG1} - y_{AG2}}{y_{AG1}} = 1 - \frac{y_{AG2}}{y_{AG1}} . \tag{13}$$

Examining the separation efficiency $\eta$, we can see from *Eqs.* (12) and (13) that the determination of $\eta$ is equivalent to the determination of the lower limit of $y_{AG2}$ from *Eq.* (12).

*Fig. 3* reveals that

$$\Delta y_A^* = y_{AG} - m x_{AL} = \left(1 - \frac{m}{\frac{L}{G}}\right) \left( \frac{\Delta y_{A2}^*}{1 - \frac{m}{\frac{L}{G}}} + y_{AG} - y_{AG2} \right) \tag{14}$$

and using *Eq.* (12), the number of transfer units is:

$$N_{OG} = \frac{1}{1 - \frac{m}{\frac{L}{G}}} \ln \frac{\Delta y^*_{A1}}{\Delta y^*_{A2}} . \tag{15}$$

Based on *Fig. 3*, we can also write that

$$\Delta y^*_{A1} = y_{AG1} - m x_{AL2} - \frac{m}{\frac{L}{G}} (y_{AG1} - y_{AG2}) \tag{16}$$

and

$$\Delta y^*_{A2} = y_{AG2} - m x_{AL2} .$$

With this and *Eqs.* (10) and (15) we obtain

$$\frac{Z}{H_{OG}} = N_{OG} = \frac{1}{1 - \frac{m}{\frac{L}{G}}} \ln \frac{y_{AG2} - m x_{AL2} - \frac{m}{\frac{L}{G}} (y_{AG1} - y_{AG2})}{y_{AG2} - m x_{AL2}} . \tag{17}$$

Replacing $\eta$ with what we know from *Eq.* (13), we get

$$\exp \frac{Z}{H_{OG}} \left(1 - \frac{m}{\frac{L}{G}}\right) = \frac{1 - \frac{m}{\frac{L}{G}} \cdot \eta - m \frac{x_{AL2}}{y_{AG1}}}{1 - \eta - m \cdot \frac{x_{AL2}}{y_{AG1}}}$$

and from here now we can calculate the actual efficiency of the separation:

$$\eta = \frac{\left[\exp \frac{Z}{H_{OG}} \left(1 - \frac{m}{\frac{L}{G}}\right) - 1\right] \left[1 - m \frac{x_{AL2}}{y_{AG1}}\right]}{\exp \frac{Z}{H_{OG}} \left(1 - \frac{m}{\frac{L}{G}}\right) - \frac{m}{\frac{L}{G}}} . \tag{18}$$

If $x_{AL2} = 0$ in the washing liquid when it enters, then *Eq.* (18) becomes

$$\eta = \frac{\exp \frac{Z}{H_{OG}} \left(1 - \frac{m}{\frac{L}{G}}\right) - 1}{\exp \frac{Z}{H_{OG}} \left(1 - \frac{m}{\frac{L}{G}}\right) - \frac{m}{\frac{L}{G}}} , \tag{19}$$

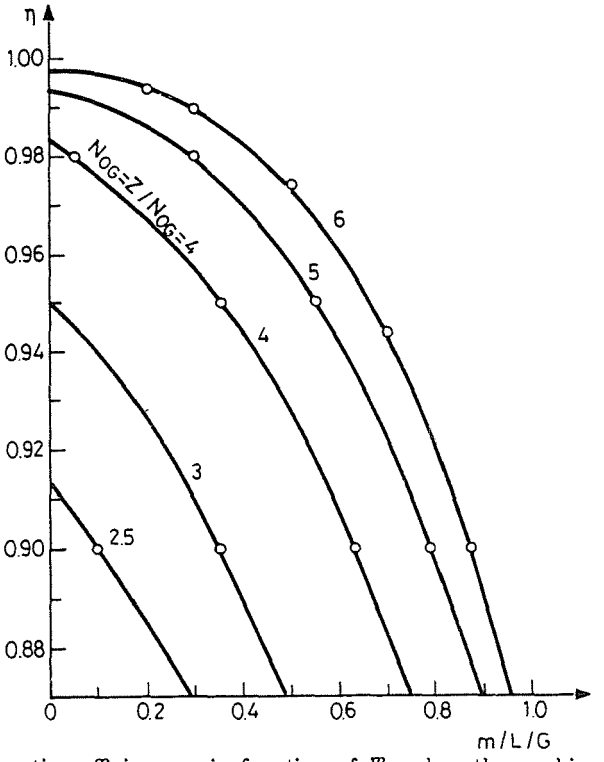


*Fig. 4.* Separation efficiency $\eta$ in function of $\frac{m}{\frac{L}{G}}$, when the washing liquid is of purity $x_{AL2} = 0$, in the case of physical absorption

which is simpler than the previously written *Eq.* (18). The variation of the efficiency $\eta$ while keeping $N_{OG} = Z/H_{OG}$ constant, is shown in *Fig. 4.* It can be seen that the greater the height $Z$, the lower the height of the transfer units $H_{OG}$, the smaller the Henry constant $m$ (that is the cooler the washing fluid), and the greater the $L/G$ ratio, *the better the efficiency of the process.* From *Fig. 4,* we can also see that $\eta = 1$ may be achieved only with a column of infinite height or zero gas flow.

Returning now to *Eq.* (18), we find that the efficiency will be lower if $x_{AL2}$ increases and/or $y_{AG1}$ decreases. The deleterious effect of the above may be partially offset by cooling the liquid, which will cause a decrease of $m$.

## Chemosorption

In the case of rapid, irreversible reactions, when component 'A' from the gas phase reacts with component 'B' from the washing liquid, then the

model of HATTA [3] assumes that the chemical reaction between 'A' and 'B' takes place at a distance of $\delta_A$ from the gas/liquid boundary layer. This place is taken to be at a distance $\delta_B$ from the bulk flow of component 'B' (see *Fig. 5*). In the place where the reaction takes place, the concentrations of 'A' and 'B' are nil, and this is where component 'C' is formed. The diffusion rate of component 'A' is

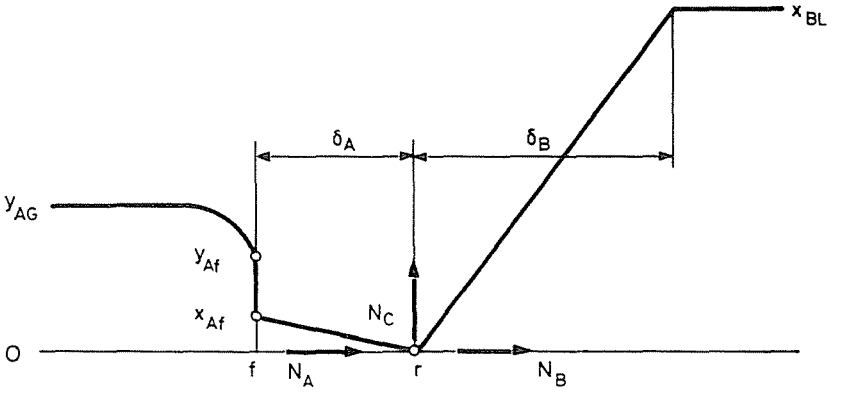


*Fig. 5.* Diffusional model of chemosorption

$$N_A = k_{AG}\,(y_{AG} - y_{Af}) = \frac{D_{AB}}{\delta_A} x_{Af} = k_{AL} x_{Af}\,, \tag{20}$$

where according to *Eq.* (3), we have again

$$y_{Af} = m x_{Af}$$

and with this

$$N_A = K_y y_{AG}\,, \tag{21}$$

where $K_y$ is the overall mass coefficient defined in *Eq.* (4)

$$K_y = \frac{1}{\dfrac{1}{k_{AG}} + \dfrac{m}{k_{AL}}}\,.$$

Following the model given in *Eq.* (9) and using *Eq.* (21), the height of the column becomes

$$Z = -\frac{G}{a}\int_{y_{AG1}}^{y_{AG2}} \frac{\mathrm{d}y_{AG}}{N_A} = -\frac{G}{K_y a}\int_{y_{AG1}}^{y_{AG2}} \frac{\mathrm{d}y_{AG}}{y_{AG}} = \frac{G}{K_y a}\left[-\ln \frac{y_{AG2}}{y_{AG1}}\right].$$

Taking into account the definition of efficiency as defined in *Eq.* (13) we get

$$Z = \frac{G}{K_y a}[-\ln(1-\eta)]\,. \tag{22}$$

Now assuming that if $G$ = constant, then in the fluid phase

$$k_{AL} = C_A L^{\Phi} \tag{23}$$

which means that it is dependent on a power of $L$, then

$$\frac{G}{K_y a} = \frac{G}{a}\left(\frac{1}{k_{AG}} + \frac{m}{C_A L^{\Phi}}\right) = \frac{G}{a k_{AG}} + \frac{G^{1-\Phi} m}{a C_A \left(\frac{L}{G}\right)^{\Phi}}\,.$$

Combining this with *Eq.* (22), we get

$$\frac{1}{-\ln(1-\eta)} = \frac{1}{Z}\left(\frac{G}{k_{AG} a} + \frac{G^{1-\Phi} m}{C_A \left(\frac{L}{G}\right)^{\Phi} a}\right)\,. \tag{24}$$

If $k_{AG}$ is not a function of $L$ but only of $G$, then in *Eq.* (24), $Z$, $G$ and $m$ are constants, then it is directly proportional to $(L/G)^{\Phi}$.

If we are cooling the washing liquid, then the Henry constant $(m)$ is also decreasing and as a consequence of this, the slope of the straight curve characterized in *Eqs.* (22) or (24) also decreases. Since this is accompanied by the decrease of $1/-\ln(1-\eta)$, it means that the separation efficiency $\eta$ improves. These conclusions are supported by our practical measurements [4] illustrated in *Figs. 6* and *7*.

However, the relation (22) alone does not express the influence of the concentration of components 'A' and 'B' or the effect of the reaction equations. Thus, we should proceed to the examination of the diffusion rate of component 'B'. The diffusion rate of component 'B' can be inferred from *Fig. 5* as being

$$N_B = k_{BL} \cdot x_{BL}\,. \tag{25}$$

The material balance on the liquid side is

$$N_B \mathrm{d}A_f = L \mathrm{d}x_{BL}\,. \tag{26}$$

From relations (6) and (8) and according to the conditions imposed by the other $(B)$ constituent, the height of the equipment is

$$Z = \frac{L}{a}\int_{x_{BL1}}^{x_{BL2}} \frac{\mathrm{d}x_{BL}}{N_B} = \frac{L}{k_{BL} a}\int_{x_{BL1}}^{x_{BL2}} \frac{\mathrm{d}x_{BL}}{x_{BL}} = \frac{L}{k_{BL} a}\left(-\ln\frac{x_{BL1}}{x_{BL2}}\right)\,. \tag{27}$$

If according to the equation of the reaction

$$N_B = bN_A \tag{28}$$

then from *Eqs.* (7), (24) and (28) we have

$$bG\,(y_{AG1} - y_{AG2}) = L\,(x_{BL2} - x_{BL1})\,. \tag{29}$$

Thus, according to the definition of $\eta$ from (13)

$$\eta = \frac{y_{AG1} - y_{AG2}}{y_{AG1}} = \frac{L}{bG}\frac{x_{BL2} - x_{BL1}}{y_{AG1}}, \tag{30}$$

that is

$$\frac{x_{BL1}}{x_{BL2}} = 1 - \frac{bG}{L}\frac{y_{AG1}}{x_{BL2}}\eta\,. \tag{31}$$

Finally, the height of the column is given by

$$Z = \frac{L}{k_{BA}a}\left[-\ln\left(1 - \frac{bG}{L}\frac{y_{AG1}}{x_{BL2}}\eta\right)\right]. \tag{32}$$

Since the height $Z$ should be identical with the calculated based on the diffusion rates of components 'A' and 'B', the combination of *Eqs.* (22) and (32) will yield

$$Z\left(\frac{K_y a}{G} + \frac{k_{BL}a}{L}\right) = \left[-\ln(1-\eta) - \ln\left(1 - \frac{bG}{L}\frac{y_{AG1}}{x_{BL2}}\eta\right)\right]. \tag{33}$$

From this

$$Z = \frac{1}{\dfrac{K_y a}{G} + \dfrac{k_{BL}a}{L}}\left\{-\ln\left[(1-\eta)\left(1 - \frac{bG}{L}\frac{y_{AG1}}{x_{BL2}}\eta\right)\right]\right\}. \tag{34}$$

The logarithm from the right side of *Eq.* (34) consists of two factors, one is $(1-\eta)$, the other is $(1-d\eta)$, where $d$ is given by

$$d = \frac{bG}{L}\frac{y_{AG1}}{x_{BL2}}\,. \tag{35}$$

Let us examine in the following *Eqs.* (3) and (4) for the specific case of the separation of sulphur dioxide from flue gases, by aqueous washing.

1 mol of sulfur dioxide (A-component) and 1 mol of water (B-component) will react:

$$SO_2 + H_2O = H_2SO_3$$

to form 1 mol of sulphureous acid (C-component).

According to *Eq.* (28), in *Eq.* (35) $b = 1$. In the case of aqueous washing $x_{BL} \cong 1$. The concentration of sulfur dioxide in smoke gases is low enough so that in *Eq.* (35) $d \ll 1$. As a consequence, $1 - d\eta$ may be regarded as a modifying factor standing beside $1 - \eta$ (with the exception of the values $\eta \cong 1$).

The modifying effect of the expression $d$ is such that the higher the concentration of sulphur dioxide in the gas is, the lower the efficiency $\eta$ becomes. These findings are supported by our results shown in *Figs. 6* and *7*, which apply to washing with water and calcium hydroxide solutions. The results of our experiments are given in [4].

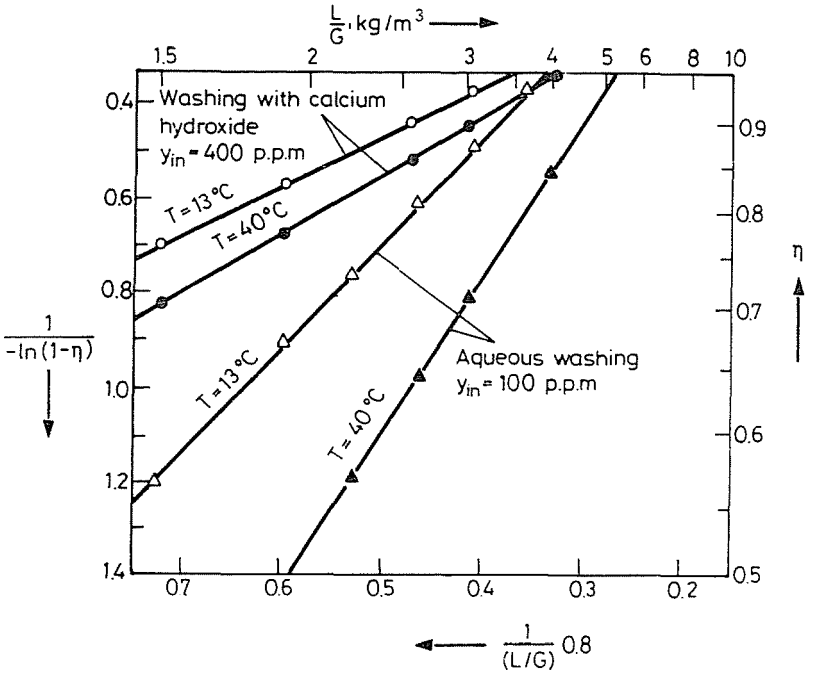


*Fig. 6.* Separation efficiency $\eta$ of sulfur dioxide in function of $\frac{L}{G}$, assuming an experimental setup of diameter $D = 130$ mm and height $Z = 350$ mm and aqueous washing and washing with calcium hydroxide solution with concentration of 27 g calcium hydroxide per kg of water, respectively, assuming a gas velocity of $v_G = 3$ m/s, a sulfur dioxide concentration of $y_{in} = 400$ ppm, the temperature of the washing liquids being $T = 13°C$ and $T = 40°C$, respectively

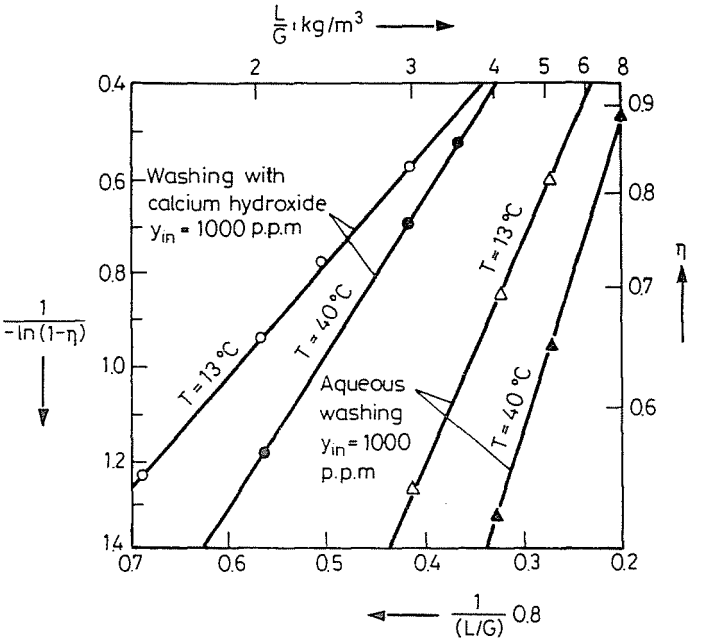


*Fig. 7.* Separation efficiency $\eta$ of sulfur dioxide, in function of $\frac{L}{G}$ assuming an experimental setup with diameter $D = 130$ mm and height $Z = 350$ mm with aqueous washing and washing with a calcium hydroxide solution containing 27 g of calcium hydroxide per kg of water, respectively, assuming a gas velocity $v_G = 3$ m/s and a sulfur dioxide concentration of 1000 ppm, the temperature of the washing liquids being $T = 13°C$ and $T = 40°C$, respectively

## *List of Notations*

| | |
|---|---|
| $A$ | surface, cross-section |
| $a$ | heat or mass transferring surface in the unit volume |
| $b = N_A/N_B$ | constant obtained from the equation describing the reaction of 'A' and 'B' to give 'C' |
| $C_A$ | dimensional constant, see *Eq.* (23) |
| $d$ | see *Eq.* (35) |
| $G$ | molar current strength on the gas side as applied to the empty cross-section of the tower |
| $H_{OG}$ | height of transfer unit |
| $k$ | mass transfer coefficient |
| $K$ | overall mass transfer coefficient |
| $L$ | molar current strength on the liquid side as applied to the empty cross-section of the tower |

| | |
|---|---|
| $m$ | Henry's constant |
| $N$ | the rate of material transfer |
| $N_{OG}$ | the number of transfer units |
| $q$ | thermal flux density |
| $x$ | mol ratio in the liquid phase |
| $y$ | mol ratio in the gas phase |
| $\Delta y^*$ | see *Eq.* (5) |
| $Z$ | height of the column |

*Greek Letters*

| | |
|---|---|
| $\eta$ | efficiency of the separation |
| $\delta$ | film thickness |
| $\Phi$ | exponent, see *Eq.* (23) |

*Indexes*

| | |
|---|---|
| $A$ | referring to component 'A' |
| $B$ | referring to component 'B' |
| $C$ | referring to the reaction product, component 'C' |
| $f$ | interphase surface |
| $G$ | referring to gas |
| $L$ | referring to liquid |
| 1 | bottom of the tower |
| 2 | top of the tower |
| $q$ | referring to the tower cross-section |

## References

1. TREYBAL, R. E.: Mass Transfer Operation. McGraw – Hill B. Co., New York, 1969.
2. SZENTGYÖRGYI, S. – MOLNÁR, K. – PARTI, M.: Transport Processes. Tankönyvkiadó, Budapest, 1986 (in Hungarian).
3. HOBLER, T.: Mass Transfer and Absorbers. Oxford Pergamon Press, 1966.
4. MOLNÁR, K.: Enhancement of Efficiency at Two-phase Systems. Report on research of OTKA, 1991 (in Hungarian).